Title: Behaviors of Impurity Elements at Beryllium Target Surface Bombarded by Alpha Particles (Commemoration Issue Dedicated to Professor Takuji Yanabu on the Occasion of his Retirement)

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Behaviors of Impurity Elements at Beryllium Target Surface Bombarded by Alpha Particles

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INTRODUCTION

In recent years, “Rutherford Backscattering Spectroscopy (RBS)” has been widely applied to various quantitative analyses of elements contained at surface of solids. Element-compositions and their depth-distributions as well as phenomena of element-diffusion, adsorption, migration and so on have been interestingly measured and discussed by simply examining the energy profiles of scattered projectiles, which are usually detected by a solid state detector (SSD) placed at a backward direction of the projectile beam. Alpha particles with MeV in energy have been mostly employed as projectiles.

In the RBS method, the mass resolution of target element is essentially better as the element becomes lighter, since backscattered alphas have less kinetic energy. Therefore elements of heavier masses contained in a light element target are easily discriminated even up to ppb order, and this has induced versatile biological and environmental applications.

The authors' group has continued the experiment of sputtering for these several years, in which sputtered heavy atoms are deposited on extra-pure carbon and the atom quantities are measured by using the RBS technique. In this experiment, trace elements presumably ascribable to a decomposition of pumping oil molecules have been observed to be inevitable. In order to examine this phenomenon precisely, it is necessary to use a lighter target such as beryllium and measure deposited atoms with and without a heavy bombardment of projectiles. To pursue them according to the history of target temperature, irradiation or baking would be important.

It has frequently been experienced by many investigators that bombardment of light ions, such as protons and alpha particles, induces a colored “beam trace” on a solid target.

KEY WORDS: beam trace/ surface oxidation and carbonization/ surface impurity/ alpha irradiation/ beryllium/ RBS/

A beryllium target has been bombarded by an energetic beam of alpha particles and the RBS measurements have been applied to know the behaviors of impurity elements, especially of carbon and oxygen, under various dose and temperature conditions. Carbon compounds initially attached are easily removed by heating up but a deposition of carbon atoms decomposed from oil vapors follows. Oxygen atoms are accumulated on a heated target, which come from decomposed water molecules. During a heavy and long bombardment, diffusion of both atoms becomes dominant and this suggests carbonization and oxidation of beam-excited beryllium atoms.
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when the vacuum chamber is evacuated with oil diffusion pump. This trace sometimes shows interference of light. This has been thought mostly attributable to deposited carbon atoms which are decomposed of oil vapors. Contributions of oxygen from existing water molecules could not be ignored. However precise observations have been poorly reported.

In this article, therefore, is stated the RBS study for trace elements of an alpha-irradiated beryllium target. The relative quantities and behaviors of these elements against dose and target temperature are described.

EXPERIMENTAL

An alpha particle beam extracted from a 4MV Van de Graaff equipment at the Department of Nuclear Engineering in Kyoto University was introduced into a 10-inch vacuum chamber and hit on a beryllium target, as shown in Fig. 1. The target was a disc of 10 mm in diameter and 1.5 mm in thickness and was set 45° with respect to the incident beam. Scattered alphas were detected with an SSD placed at 135° backwards and the energy spectrum was displayed on a 512 channel pulse height analyser.

In order to know the temperature effect, the beryllium target was mounted on an assembly of an insulator and a tantalum heater, the temperature of which being measured by the use of a chromel-alumel thermocouple. A system of a 6″ oil diffusion pump and a liquid nitrogen trap served to evacuate the experimental chamber. The base pressure was 1×10⁻⁶ Torr. The alpha beam was about 2 mm in size, the energy and intensity of which being 1.6 MeV and about 1 μA throughout the irradiation, respectively.

The RBS observations were carried out for a fixed beryllium target under various conditions—fresh, heating up, and beam irradiations at RT, 280, 50, 340, and 450°C. From the alpha spectra collected at these stages, valuable informations concerning the trace elements were drawn out.

The analyses of elements were carried out according to the procedures described in the texts.\textsuperscript{23}

The thickness of \( d \) of the surface layer is approximately given by

\[ d = \frac{\Delta E}{S(E_0, k)}, \tag{1} \]

where \( E_0, \Delta E, \) and \( S(E_0, k) \) represent the incident energy, the energy spread in the spectrum, and the energy loss factor for the backscattered particles, respectively.
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being the so-called backscattering coefficient. The atomic density $N_A$ of atom A at the surface layer can be thus given as

$$N_A = \frac{\sigma_{Be}/\sigma_A}{\langle S(E_{b1}, k_A)/S(E_{b1}, k_{Be}) \rangle} \langle C_A/C_{Be} \rangle N_{Be},$$

where $\sigma$ and $C$ denote the Rutherford scattering cross-section and the counts per unit energy interval, respectively. Notations A and Be correspond to atoms A and Be.

RESULTS AND DISCUSSION

The obtained pulse height spectra are compared each other by normalizing continuum yields due to beryllium, as shown in the next figures of energy profiles. This is the simplest way to know the behaviors of trace elements under various circumstances.

1. Virgin target (room temperature)

The backscattered alpha spectrum from a virgin target is demonstrated in (a) of Fig. 2. Carbon and oxygen are found considerably dominant and in particular, the carbon peak is sharp enough. Considering the energy loss of impinging alphas within

![Energy spectra of backscattered alpha particles for a fixed beryllium target under various conditions: (a) virgin target at RT, (b) after 20 min at 280°C without irradiation, (c) after 30 min irradiation at 50°C, (d) after 105 min irradiation at 50°C.](image)
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the target, the depth of carbon atoms is equivalent to $\Delta E=24\,\text{keV}$ FWHM according to Eq. (1), corresponding to 1700 Å. This implies that carbon molecules or carbon atoms are thinly distributed at the surface. The oxygen peak gives about a 28 keV spread, that is 1500 Å, but its tail to low energy side means a deeper distribution.

Trace elements of fluorine, sulphur, chlorine, calcium and those of masses ≈60 are also existing. The reason cannot be simply discussed at present, but the contamination in manufacturing processes is considered.

2. Preheating (280°C - 20 min)

When the target was preheated 20 min at 280°C, almost trace elements other than oxygen disappeared, as shown in (b) of the same figure. This tells that these elements are loosely attached to the beryllium target as considered above and carbon compounds, if used, are easily escaping or decomposable.

In contrast to these, the oxygen peak drastically increased up to a few times than before. The peak profile suggests that oxygen atoms still stay at the target surface, thus we deduce that water molecules are decomposed on heated beryllium and its oxidation proceeds.

3. Alpha irradiation (50°C -30 min, -105 min)

The preheated target was cooled down and the alpha irradiation of 30 minutes was done. Since the beam power was about 1.6 watts ($\sim 1\,\mu\text{A} \times 1.6\,\text{MeV}$), the target temperature increased up to 50°C.

The observed alpha spectrum is depicted in (c) of Fig. 2. The peak yield of oxygen does not so differ from (b), but the carbon peak together with the base yield (counts of 70-170 channels) grows up considerably. This means that surface oxidation of beryllium is almost steady, while another process of carbon contamination starts also at the surface. On the other hand, carbon and oxygen atoms begin to move in the target, which results in an increase of the base counts.

These behaviors can be more clearly understood when the irradiation is continued up to 105 minutes. The result is compared in (d), where the carbon yield becomes more dominant—approximately twice of (c).

4. Irradiation at a raised temperature (340°C -40 min)

The target temperature was then raised to 340°C and the beam irradiation was continued. The alpha profile after 40 minutes is demonstrated in (e) of Fig. 3, in which broadening of carbon and oxygen peaks appears. This implies that the surface densities of these atoms do not change seriously but diffusion into the target interior becomes significant. The depths are estimated to be 3000 and 2700 Å for carbon and oxygen atoms, respectively.

5. No treatment in vacuum (20°C -500 min)

When the target was left 500 minutes in the vacuum chamber with no treatment, the RBS pattern showed a slight increase of carbon atoms whereas the oxygen yield did not show any meaningful change, as seen in (f) of the figure. This predicts that
residual gas molecules containing carbon are more adsorbable onto beryllium specimen at room temperature, whereas oxygen atoms do at high temperature. Beryllium oxidation resulting from a heat decomposition of existing water molecules seems again plausible.

6. Re-irradiation at an increased temperature (450°C —20 min)

The beam irradiation was repeated at 450°C and the RBS examination was inserted every 10 minutes. The carbon peak decreased at first but the recovering accumulation together with diffusion began, while the oxygen peak became broader. These phenomena are quite similar to those in the former stages of (b)–(d).

A representative profile after 20 minutes treatment is demonstrated in (g) of Fig. 3, where both carbon and oxygen atoms are more dense and more widely distributed. The calculated depths reach 4200 and 4700 Å for carbon and oxygen atoms, respectively.

7. Long irradiation (≥450°C —20 min)

The bombardment was continued long at 450°C or more. In (h) of Fig. 3 is represented the spectrum after 200 minutes irradiation, where heavy accumulation and diffusion of carbon and oxygen atoms are seen. Their depths are crudely estimated to be 10000 Å or more—a few times thicker than the wave length of visible light. A brownish “beam trace” appears at this stage. At the surface layer the atomic densities of carbon
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and oxygen, relative to that of beryllium, are estimated according to Eq. (2) to be 0.2 and 0.6, respectively. A little adsorption of silicon atoms becomes distinguishable, which can be ascribed to a decomposition of silicon oil vapors.

Throughout the present RBS observations of alpha irradiated beryllium target, the following conclusions are derived:

1. A removal of simply attached carbon compounds can be attained by a heating-up procedure for the target. As the beam irradiation proceeds, however, carbon atoms decomposed from oil vapors are deposited on the target surface and make a diffusion according to the target temperature. Carbonization of beryllium would take place by the action of impinged alpha energy.

2. An adsorption of oxygen does not proceed at the room temperature. When the target is heated up to a few or several hundreds °C, oxygen atoms ascribable to a decomposition of water molecules are found. These atoms diffuse into the interior more easily than carbon atoms do. This means that oxidation of beryllium is rather dependent on the temperature and proceeds swifter than carbonization.

3. The depth of carbon atom increases as the alpha fluence becomes larger. It is about 1500 Å at the initial stage, but exceeds 10000 Å after the irradiation of 1μA × 400 min. A brownish spot is seen at the irradiated position, thus the so-called "beam trace" can be clearly attributed to carbon atoms decomposed from oil vapors.

4. Oxidation or carbonization is dependent on target element, its environment and temperature, thus the "beam trace" cannot be quantitatively discussed for all targets. Even in an oil-free and ultra high vacuum, the chemical and physical behaviors of impurity elements would be complicated because of their excitation by projectiles. It should also be noted that a thick oxide layer would be formed on active metals, provided that its temperature is raised in a vacuum system where water molecules considerably exist.

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